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Behavior of plutonium isotopes in the marine environment of Enewetak atoll

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There continue to be reports in the literature that suggest a difference in the behavior of $^{239+240}$Pu and ^{238}Pu in some aquatic environments. Plutonium isotopes have been measured in marine samples collected over 3 decades from Enewetak atoll, one of the sites in the Marshall Islands used by the United States between 1946 and 1958 to test nuclear devices. The plutonium isotopes originated from a variety of complex sources and could possibly coexist in this environment as different physical-chemical species. However results indicate little difference in the mobility and biological availability of $^{239+240}$Pu and ^{238}Pu.

Introduction

Natural processes in the environment are unlikely to differentiate among the plutonium isotopes, but there have been and continue to be reports in the literature suggesting ^{238}Pu in some marine environments is more mobile and biologically available than $^{239+240}$Pu.[1–4] In some cases the reason for the difference in behavior is not altogether clear. The investigators reporting these findings appear to have considered the often discussed problems[1,3,5,6] related to possible sources of error from sample collection, analytical separations and detection of plutonium isotopes. Differences in behavior can also arise if one isotope is present in an environmental media as a different physical-chemical species.[3] The plutonium isotopes could exist in different physical-chemical forms at Enewetak Atoll. Enewetak was one of the sites in the Pacific used by the United States between 1946 and 1958 to test a number of nuclear devices. The plutonium isotopes that were generated during testing were deposited to the local environment. Since 1958 the sedimentary reservoir of the lagoon has been source term for plutonium and other radionuclides. Quantities of the radionuclides are now remobilized (involving loss by desorption or some other means), resuspended, assimilated, and transferred continuously within the atoll environment by physical, chemical and biological processes. Past aquatic studies at Enewetak were related to better understanding $^{239+240}$Pu behavior in the environment and to assess the contribution of effective radiation dose to humans from $^{239+240}$Pu in marine pathways.[7–15] Much of the ^{238}Pu data that was measured concurrently with $^{239+240}$Pu has not been previously published. A comparison is now made of the relative concentrations of ^{238}Pu and $^{239+240}$Pu to assess if significant differences in behavior are evident among these isotopes in this atoll environment.

Experimental

The results in this report were generated from samples collected over the last 3 decades. Field and laboratory personnel changed during this period but sample collection methods, processing and radiochemical procedures were not significantly altered. Therefore, previously published documents adequately describe field collections of sediment, water, and different biota from Enewetak (and other Marshall Island Atolls) and the analytical techniques used at Lawrence Livermore National Laboratory for the separation and analysis of plutonium isotopes by alpha spectrometry.[6–16] It was necessary to correct all ^{238}Pu ($T_{1/2}$ = 87.74 y) concentrations for decay to a common date, arbitrarily taken as 1 January 1997, to better compare results collected over the past 23 years. The measurement of ^{238}Pu was a byproduct from the analysis of larger quantities of $^{239+240}$Pu by alpha-spectrometry and subjected to larger errors. In many cases, especially for water and fish samples, the ^{238}Pu was below limits of detection because of insufficient sample size, an inadequate counting period, low chemical recovery or a combination of these factors. ^{242}Pu was added to dissolved samples to determine recoveries of plutonium. The quality of our analytical results was continually checked over the years through analysis of sample duplicates and blanks. Data reliability was confirmed through regular participation in national and international intercomparison exercises.

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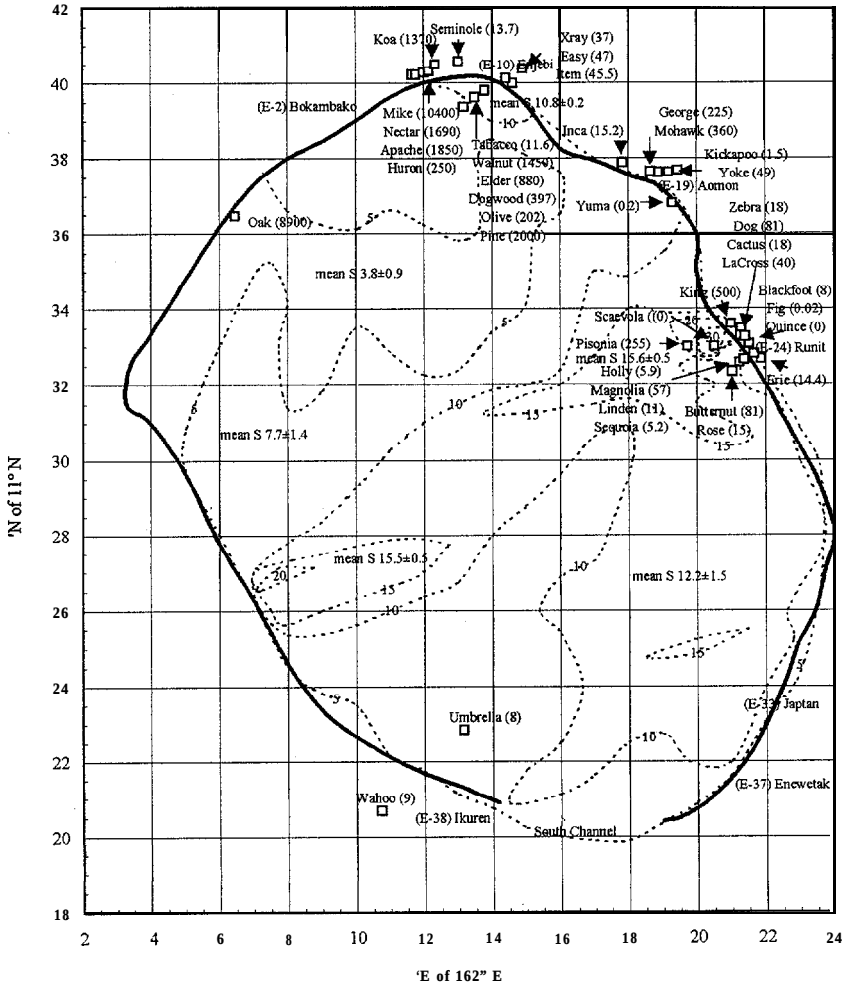

Fig. 1. Outline of Enewetak atoll with nuclear test names, locations and yields (in parenthesis). Contours for S values [see text and Eq. (I)] associated with lagoon surface sediments are shown. S values are given as of 1 January 1997. Islands where reef fish were caught for analysis are identified by name and code number (in parenthesis)

Different dimensionless quotients have been used in the literature to express the relative concentrations of the alpha emitting isotopes of plutonium such as the ^{238}Pu:$^{239+240}Pu$ ratio; the $^{239+240}Pu$:^{238}Pu ratio; and these values expressed as percentage. In this report all relative concentrations are expressed as an S value. The dimensionless value is defined by Eq. (1), expressed as a percentage, to describe the activity of ^{238}Pu relative to the alpha emitting isotopes of plutonium.

$$S = (^{238}\text{Pu alpha activity}/(^{238}\text{Pu} + ^{239}\text{Pu} + ^{240}\text{Pu alpha activity})) \times 100 \quad (1)$$

In samples where ^{238}Pu was below detection limits, S values were not computed.

Results and discussion

Nuclear testing and plutonium sources

The names, locations, and kiloton yields (shown in parentheses) of the 43 different tests conducted at Enewetak atoll are shown in Fig. 1. Nineteen of the tests were exploded on barges anchored in the lagoon. The remaining tests consisted of 2 air drops, 2 underwater explosions, 7 ground surface tests, and 13 where devices were fixed to towers. Most of the large yield devices were exploded in the northwest quadrant of the lagoon, but the largest number of tests was held near Runit Island (shown in Fig. 1) on the eastern reef of the atoll. The following demonstrates that the plutonium isotopes in the environment originated from many different source terms during the testing program.

During the 1958 Scaevola and Quince tests, only the high explosive components of the devices were detonated. Plutonium nuclear fuel (probably as oxides) was scattered over the nearby region. The decay corrected S value for this oxide and other sources of weapon-grade plutonium, estimated from samples collected at accident sites in the marine environment, range from 1.5 to 1.9 (1.8 to 2.2 on the date of collection).[17,18]

Quantities of ^{239}Pu, unassociated with any ^{238}Pu, can be produced by capture reactions and parent decay. It is estimated that about 67% of the total Pu deposited on the surface of the earth as global fallout was produced by the (n,γ) reaction with ^{238}U and the subsequent decay of ^{239}U to ^{239}Np to ^{239}Pu.[19] Therefore, this mode of production generated ^{239}Pu as single atoms some time after the explosions. In other events, such as those during the 1948 operation Sandstone at Enewetak, only about 35% of the plutonium[20] in the devices was utilized in fission. The remaining was most likely deposited to the local environment as small particles of ^{239}Pu oxide.

An S value[21] of 0.6 was associated with the close-in fallout material from the "Mike" thermonuclear test in 1952 at Enewetak. We recently determined a value of 0.10 (8% error) measured in debris that fell on the Japanese fishing vessel, Fukuryu-Maru, from the 15 Mt Bravo explosion at Bikini in 1954. Thus the larger thermonuclear tests generate small relative amounts of ^{238}Pu.

An S value greater than 30 is associated with sediments from Cactus crater on Runit Island. This crater and associated debris resulted from a single 18-kt explosion during the Hardtack I test series on 5 May 1958. This is an exceptionally high S value and would require that ^{238}Pu be produced from decay of an unreasonably large amount of a parent radionuclide (such as ^{242}Cm, or ^{238}Np) or generated from some special material associated with the device. Even the production of ^{240}Pu differs from that of both ^{238}Pu and ^{239}Pu so that based on differences in production mechanisms alone, the characteristics of the three alpha emitting isotopes in regions within this atoll environment could differ greatly.

Plutonium isotopes in lagoon sediments and seawater

The concentrations of $^{239+240}Pu$ and ^{238}Pu were determined in 200 samples of surface sediment and in vertical sections of several core samples collected from all regions within the lagoon during the 1970's. The lagoon sediments are heterogeneous and composed primarily of different quantities of fine-and coarse-grained carbonate material, remains of shells, coral fragments, and *Halimeda* (calcareous algae) debris and foraminifera.[22] In over 98% of the sediment samples the $^{239+240}Pu$ concentration associated with the fine sediment components (<0.5 mm) was greater than or equal to the concentration associated with the coarse fraction. The plutonium isotopes are also nonuniformly distributed aerially and vertically in the lagoon sediment.[10] All regions of the lagoon received some amount of plutonium isotopes from the different tests. The highest concentrations of $^{239+240}Pu$ in surface sediments[7] are in the northwest quadrant of the atoll in a north and south oriented elliptical area several kilometers southwest of the Mike and Koa test site shown in Fig. 1. There is also a relatively high $^{239+240}Pu$ contaminated area of sediment extending southwest of Runit island.[7] Regions of the lagoon with similar S values (decay corrected) in the surface sediment are shown in Fig. 1. Mean values of S (with standard deviations) within each contour are also shown. The computed mean value and standard deviation for S using all 200 data points for the entire lagoon is 9.8±4.5. This is approximately a factor of 4 greater than the (decay corrected) S value associated with global fallout and SNAP debris delivered to the latitudes of the Northern Marshall Islands.[21]

Figure 1 demonstrates the complex distribution of the plutonium isotopes in components of the surface sediment. S values range from <2 in sediment samples from the west-northwest region of the Atoll to 31 in material from Cactus crater. The isotopes were not introduced in the same relative proportions but varied during the period of testing. The differences in S values relate to specific events, not to processes that might lead to discrimination among the isotopes.

Values of S associated with sediment in sections of core samples are frequently irregular and are found to increase or decrease in value, or can be constant with depth. For example, material collected shortly after the 1952 Mike event had an S value of 0.6.[21] Values of S in sections of sediment core samples from Mike crater now range from 7.5 at the surface to 4.1 at a depth of 50 cm. Thus a mixture of material originating from the post 1952 tests, labeled with different S values, settled in the crater over time to alter the original value of 0.6. On the other hand, the S values found in Cactus crater sediment were constant along the length of sediment core samples indicating that only a single source labeled the sediment in this crater.

$^{239+240}Pu$ concentrations and S values associated with seawater and components separated from sediments collected at two mid-lagoon locations and in Cactus crater are shown in Table 1. The plutonium isotopes have not remained permanently fixed to the fine sedimentary material but are readily exchanged among the living and inactive components in bottom sediments. In addition, to within our analytical precision, the value of S associated with the solid and liquid phases at the two mid-lagoon locations are identical. Results from Cactus crater are particularly relevant to the argument of

possible discrimination. Recall that the ^{238}Pu associated with material from this crater could have originated from a source unrelated to $^{239+240}Pu$. Nearly identical values of S are found in the interstitial water, filtered crater water, particulates from different depths, fine and coarse sediment, and live algae. The values in Table 1 for water are mean concentrations determined in samples collected during different tidal stages over a 5 year period. Values of S were essentially identical in the water samples during the 5-year period. The relative rate of mobilization for the ^{238}Pu and $^{239+240}Pu$ isotopes from the sedimentary source in the crater to the water column was unchanged with time. These examples demonstrate essentially no discrimination among plutonium isotopes in the release or exchange from sedimentary source terms to bottom water and in the subsequent accumulation by living and inanimate sedimentary components over time.

A large number of seawater samples have been collected from all regions of the lagoon over the past 3 decades (22 years). Most samples were filtered and the plutonium concentration was determined in the filtered solution and sometimes in the particulate fraction removed with 1-μm filters. A one-micron filter was found to be equally efficient as a 0.45 and/or 0.2 μm filter in removing particulates from the seawater.[23] Values for S in the filtered seawater ranged from 1.7 to 20. This is also essentially the range in S values found associated with the lagoon sediments. Therefore, no one region of the atoll is singly contributing the radionuclides to the water column. Plutonium isotopes are continuously mobilized to solution everywhere as the seawater flows in contact with the sediments or with materials on the reef.

The mean (decay corrected) S value in solution from all filtered water samples is 9.5±2.9 and the mean value found associated with the particulate samples is 10.0± 5.9. There is no significant difference between the average S value found in seawater solution and associated with the particulate material.

During 1972, 74, 76, 82 and 1994, the mean values for S associated with the water samples were 9.6±2.2, 9.2±2.6, 9.4±2.7, 8.2±1.8 and 13.8±6.1, respectively. A different number of samples were analyzed each year but the mean did not change significantly (within experimental error) over time. Reduced and oxidized concentrations of the plutonium isotopes were determined in 18 water samples from different lagoon locations. Separation of the oxidation states involved a double tracer technique. The mean S value found associated with the oxidized and reduced forms were 9.5 ±0.8 and 10.1±3.1, respectively. There appeared to be no preferential enrichment of either isotope as a soluble reduced or oxidized species. The mobilized forms partition equally among oxidation states in the water mass where 79±6% of the total ^{238}Pu and the total $^{239+240}Pu$ was found in the oxidized (+V,VI) state.

Plutonium in tissues of 3 species of reef fish from several islands of Enewetak

About 2000 fish have been collected from Enewetak Atoll for radionuclide analysis between 1972 and 1995.[24] Separated tissues and organs from each fish of a species caught at a fishing site are pooled for analysis. A final pooled sample may consist of tissues or organs from as many as 50 fish of the same species. The log normal mean (and mean value) concentration of $^{239+240}Pu$ in fresh fillets (muscle tissue) from all reef and pelagic fish collected at the atoll between 1976 and 1995 was ~0.011 Bq/kg. We limit our discussion here to the concentrations of plutonium isotopes measured in 3 reef species with somewhat different feeding habits. The fish are the mullet, a trophic level II species (*Crenimugil crenilabis* and *Neomyxus chaptalii*); Convict surgeonfish, a trophic level II species (*Acanthurus triostegus*); and Goatfish, a trophic level III species (*Mulloidichthys samoensis*). Mullet generally accumulate more plutonium in the flesh than the other two species and body burdens of the 3 species are derived both from the water and material ingested.[25] These fish are preferred in the Marshallese diet so concentration data are important to evaluate internal exposure from the marine food chain. Most of these fish spend their lifetime feeding over a relatively small territory on the reef and are easily caught using a throw net when and where they were sighted in the surf. Therefore, the fish may have been collected from different regions of an island and some variability in radionuclide concentration related to geography could be anticipated.

Mean values of S in pooled parts of fish, collected between 1976 and 1995 from the islands identified in Fig. 1, are summarized in Table 2. Mean values are provided for fillets; stomach contents and viscera; and the remaining tissues and organs that include bone, liver, skin, reproductive organs, gills, eyes separated from some but not all samples. Viscera samples also contain ingested material that often includes some sediment (especially true for mullet). At island E-24 (Runit) sufficient data were available to average the S values associated with the individual species. Water was also sampled from the Runit lagoon reef in the region where fish were caught. Some variation in S is noted in the water and gut (sediment) samples from mullet and surgeonfish. Therefore, fish from even a defined region of the reef may be exposed over time to water, food and sediment with different S values. However, over the long term the mean values found for S in the flesh, water, ingested material and other parts of the fish are very

Table 1. $^{239+240}Pu$ concentrations and S values associated with sediment components and water at two lagoon locations and in Cactus crater

Component	Station location: 162°8.37'E 11°38.11'N $^{239+240}Pu$	S value as of 1/97	162°5.14'E 11°33.1'N $^{239+240}Pu$	S value as of 1/97	162°21.25'E 11°33.28'N Cactus Crater $^{239+240}Pu$	S value as of 1/97
Mollusk shells[a]			24 ± 2	6.8 ± 2.2		
Halimeda fragments[a]	222 ± 15	6.1 ± 0.8	178 ± 18	6.1 ± 0.8		
Coral fragments[a]	370 ± 120	9.5 ± 4.2	48 ± 4	6.1 ± 2.2		
Forams[a]	100 ± 7	7.5 ± 1.5	41 ± 4	5.4 ± 2.3		
Sediment fines (0.5 mm)"	870 ± 7	7.5 ± 0.7	254 ± 2	5.4 ± 0.8	3040 ± 74	29.2 ± 1.6
Filtered bottom water[b]	1.64 ± 0.06	7.5 ± 0.6	1.20 ± 0.11	6.4 ± 1.3	3.6 ± 1.7[c]	28.4 ± 1.2
Filtered bottom particulates[b]					16.1 ± 8.6[c]	28.8 ± 1.6
Filtered mid-depth water[b]					3.2 ± 0.1[c]	27.3 ± 0.9
Filtered mid-depth particulates[b]					4.2 ± 1.3[c]	28.1 ± 1.2
Filtered surface water[b]					2.6 ± 0.7[c]	26.5 ± 1.8
Filtered surface particulates[b]					4.0 ± 2.1[c]	27.8 ± 1.9
Interstitial water[b]					9.89 ± 0.5	30.8 ± 1.3
Live *Halimeda monile*[a]					692 ± 13	29.2 ± 1.6
Sediment (tines O-2 cm)[a]					3852 ± 80	29.9 ± 0.9
Sediment (coarse O-2 cm)[a]					1378 ± 35	29.9 ± 0.9
Sediment (fines 4-6 cm)[a]					3259 ± 70	27.2 ± 2.7
Sediment (coarse 4-6 cm)"					2456 ± 60	30.6 ± 2.4
Sediment (fines 10–14 cm)"					4059 ± 80	29.7 ± 1.5
Sediment (coarse 10-14 cm)[a]					2819 ± 60	31.1 ± 2.3

[a] Bq/kg dry weight.
[b] mBq/kg wet weight.
[c] Mean concentrations and S values in filtered water and particulate samples.

Table 2. Mean values of S associated with parts of 3 reef fish from several islands of Enewetak atoll

Island ID and fish	Mean S value: Muscle (fillets) only	Stomach contents-viscera[d]	Other parts[a]	All parts
E-24[b]				
mullet	15.3±1.4	16.1fl.6	11.2±5.2 (5)	
Surgeonfish	16.4±4.3	16.3±5.1	12.3±4.0 (7)	
Goatfish	19.3±8.8			
All fish	16.7±5.1 (24)[c]	16.2±2.9 (9)	11.9±4.4 (12)	14.9±5.5(45)
Reef water (1975-94) from fishing area				14.5±4.8(12)
E-2				
All fish	6.3±2.9 (6)	4.7f1.2 (7)	6.9fl.3 (17)	6.2±1.9 (30)
E-10				
All fish	5.7f3.6 (7)	5.5±2.7 (6)	3.8f2.2 (10)	4.8±2.9 (23)
E-19				
All fish	3.2±1.1 (5)	2.7±0.3 (5)	3.3±1.2 (16)	3.1fl.l (26)
E33,37,38				
All fish	6.1±1.1 (2)	7.4±1.1 (7)	8.8±3.0 (24)	8.3±2.7 (33)

[a] Consists of values for bone, liver, skin, gills, eyes in some but not all fish.
[b] See Fig. 1 for island location.
[c] Number of samples are in parenthesis.
[d] Viscera may include large and small intestines with contents, stomach wall, spleen, kidney and mesenteries. The intestines often contain quantities of bottom sediment. Little difference was found in S values of stomach contents and viscera. Individual values were therefore combined.

similar. There is no significant enrichment of ^{238}Pu relative to $^{239+240}Pu$ in fish over the relative amounts found in the local environments. At island E-24 there was little difference found in the mean value for S among the parts of the different species. Therefore, it was appropriate to combine values of S for all 3 species at this and the other islands sampled where a more limited amount of data were available for comparison. Mean values of S in flesh and other parts of fish from all islands sampled are found to range from ~3 at island E-19 to 15 at island E-24. The different S values show these territorial feeders are exposed to different local sources of plutonium isotopes. Similar S values are found associated with all parts of the fish from the other locations sampled. There is no significant long-term enrichment of ^{238}Pu relative to $^{239+240}Pu$ in parts of fish above that found in local seawater or ingested material from any region within the atoll.

Conclusions

Different physical-chemical forms of plutonium isotopes may exist in regions of the marine environment at Enewetak Atoll suggesting that ^{238}Pu, for example, could be more biologically available to indigenous marine species than $^{239+240}Pu$. A substantial data base was developed for concentrations of plutonium isotopes in sediment, seawater, and biota collected from the atoll during the last 3 decades. An examination of all results provides no support to the argument that ^{238}Pu is more mobile in the environment or biologically available to organisms than $^{239+240}Pu$. Over time the ^{238}Pu and $^{239+240}Pu$ have been released to solution from the bottom sediments at comparable rates and are accumulated, with no preferential enrichment, by abiotic and biotic constituents in the lagoon. The isotopes partition equally between oxidized and reduced states in seawater solution. Relative concentrations of ^{238}Pu and $^{239+240}Pu$ in species of reef fish are no different than those found in the water or food (or sediment) from the local reef environment where the fish feed.

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